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The Interaction of Water and Aerosols in the Marine Boundary Layer: A Study of Selected Processes Impacting Radiative Transfer and Cloudiness

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LONG-TERM GOALS

The overarching, long-term goal of the study is to explore the profound effect of aerosol-water interaction both on radiation propagation in, and the thermodynamic structure of, the marine boundary layer. Specific goals are: 1) compile a climatology of aerosol hygroscopicity for use in the NAAPS and COAMPS models, and, further, to develop a model parameterization of hygroscopicity based on aerosol size and composition for such models, 2) explore the relative impacts of cross-inversion mixing and sub-cloud aerosol on cloud thickness and cloud base height, 3) quantify and parameterize the impact of precipitation scavenging on below cloud radiative transfer and cloud liquid water path. The sampling platform utilized is the CIRPAS Twin Otter research aircraft and the venue is the littoral environment off the California coast, representative of areas with high shipping densities.

OBJECTIVES

For the current reporting period, our efforts have centered on completing several different analyses utilizing data both from the various CARMA studies, the VOCALS study and additional studies such as SAFARI and DYCOMS, the precise suite of data bases being dependent on the specific analysis. Our objectives for these analyses have changed from those in our original proposal several times, guided by our ongoing analysis. We summarize them for the current reporting period as follows.

- Determine the relationship between cloud drop number concentration (CDNC) and the properties of the precursor aerosol that have the most prognostic power using data from the most important cloud venues from the standpoint of cloud radiative climate forcing.

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- Explore various modifications to the aerosol/chemistry portion of the NAAPS model to assess which changes would most enhance the prognostic power of the model for Aerosol Optical Depth (AOD).

- Develop a climatology of aerosol optical properties based on data from the CARMA field studies and demonstrate the utility of this data for addressing various issues of importance for estimating the impact of anthropogenic aerosols on climate..

APPROACH

The first objective involves data on CDNC and the various properties of precursor aerosols most indicative of cloud drop activation. As we have reported in the previous annual report, the obvious proxy for CDNC would be CCN concentrations and these are in fact measured with one of two instruments. The first and most widely used is the DMT CCN-100 spectrometer while the second is the University of Wyoming's MA-100 static diffusion chamber. However, CCN actually have little prognostic power without accompanying measurements of cloud supersaturation, a parameter that cannot be directly measured. It has been proposed that essentially aerosol size alone can more simply be used to predict CDNC (cf., Dusek et al, 2006). While this has been challenged (e.g., Hudson, 2007) and is likely not universally true, it may well be true for the three main stratocumulus decks of the world (off the coasts of California, Chile and Namibia, respectively), that largely control aerosol indirect forcing. Previous work has in fact suggested that the accumulation mode number concentration (AMNC) is a useful indicator of cloud drop number concentrations in such venues (e.g., Hegg et al, 2010). We have carried out an analysis of the relationship between AMNC, as measured by a PMS/DMT PCASP-100x, and CDNC, for the above three venues by means of regression analysis.

The second objective has been addressed by several different routes. First, as reported previously, comparison of measured and modeled deposition of both precipitation and various chemicals in that precipitation has been undertaken for the arctic. Secondly, several sensitivity runs with the NAAPS model have been made to assess the extent of prognostic gain for various plausible input changes. Finally, based in part on the results from these two analyses, an extensive literature review has been done to determine the best approach to incorporating aerosol organics into the model

The final objective, the development of a climatology of aerosol optical properties is addressed in a straightforward manner using standard statistical techniques to derive the distributions of the various optical properties over the various field campaigns for which we have data. Several analyses of the value of this data base in addressing important aerosol impact questions have been done.

WORK COMPLETED

To date, work on the first objective has been completed and a manuscript has been published in the journal, Atmospheric Chemistry and Physics. Work on the second objective has been completed and the results relayed to our colleagues at NRL Monterey (who operate the NAAPS model) for their consideration. This was done primarily in the form of a formal presentation at the ICAP meeting in Frascati this past spring. Work on the third objective has proceeded slowly, as discussed in previous reports, but some analysis and results are now in hand. We anticipate completing the analysis by the end of the grant period.

RESULTS

For the regression analysis of the CDNC-aerosol relationship, data were derived from two years of CARMA data (2005, 2007), and from the VOCALS-Rex study, associated with the stratocumulus decks of California and Chile, respectively. Additionally, data from the DYCOMS experiment II (Twohy et al, 2005), located in essentially the same operational area as the CARMA studies, has now been included in the analysis. While no data gathered directly by us were available for the third of the main global stratocumulus decks, that off of the coast of Namibia, a limited amount of data was gleaned from either the literature (e.g., Kiel and Haywood, 2003) or from public data archives (the archive for SAFARI 2000 data in the CARG archive at the University of Washington). For a direct comparison of CDNC with CCN active at various supersaturations (0.2 to 1.0 %), no relationship with an R^2 in excess of 0.33 could be found. However, for the CDNC-AMNC relationship, an excellent regression fit was found and is shown in Figure 1. The R^2 of 0.91 exceeds anything of which we are aware from previous work. It suggests that AMNC is a very powerful prognostic parameter for the venues examined and could provide a very useful tool both in remote retrieval of effective CCN and for use in large scale climate models.

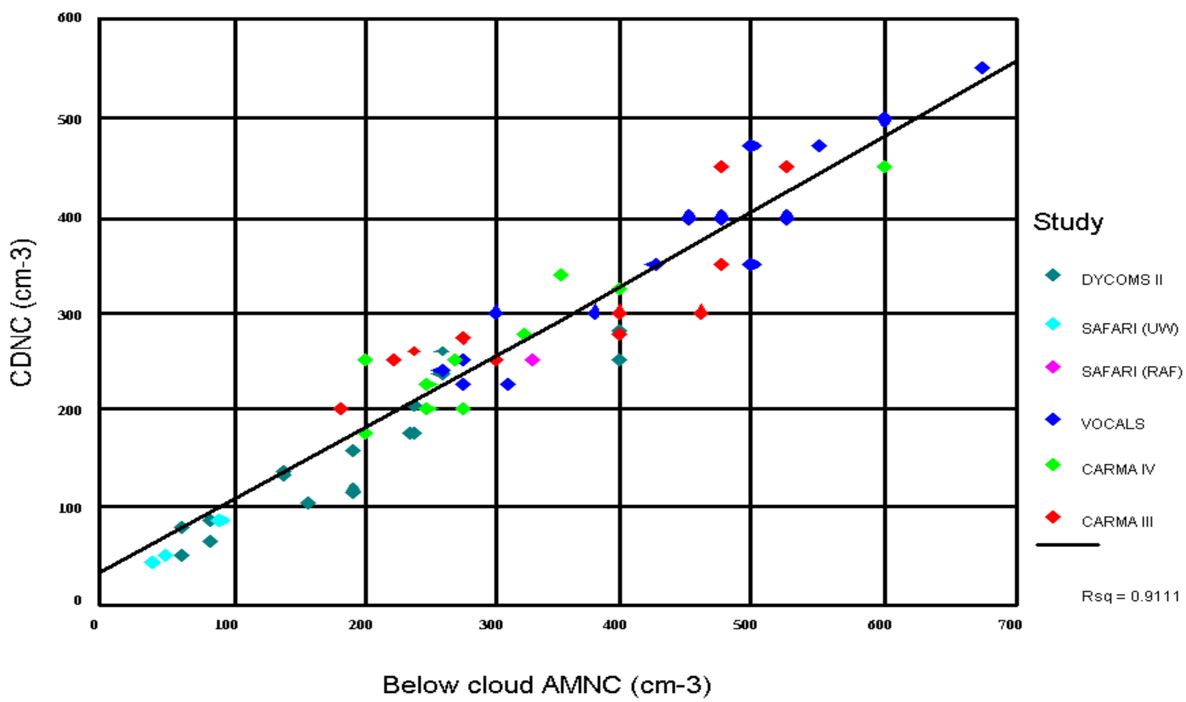

Figure 1 Regression analysis of the dependence of the CDNC on the AMNC for the three main stratocumulus decks of the earth system.

The assessment of approaches to improve prognostic power of the NAAPS model for AOD has followed several routes. As discussed in the previous annual report, it was initially thought that the relatively primitive parameterization of sulfur chemistry in the model was leading to the observed underprediction of AOD. However, a comparison measured and model predicted precipitation and sulfate deposition in the arctic did not reveal any systematic low bias in the deposition of sulfate. Furthermore, model sensitivity studies showed that varying the sulfate precursor input or model oxidation within any reasonable bounds could not correct for the aerosol mass underprediction. For

example, shown in Figure 2 is a comparison of a base model run with one in which the input of SO_2 has been doubled and essentially instantly oxidized to sulfate. Clearly the column mean sulfate concentration has not doubled. In fact, the annual average global mean column burden has increased by only ~ 35%. Since the doubling of SO_2 is a highly implausible scenario, this result, coupled with the previous reasonable agreement of model/observed sulfate deposition, render it quite clear that the low model bias for AOD is not due to sulfate chemistry. It in fact almost certainly resides in the lack of organic aerosol in the model. To address this issue, an extensive literature survey has been undertaken of the impact of organic aerosols in global models. Briefly, the results indicate that most of the organic aerosol is secondary and is of biogenic origin. With respect to the chemistry necessary to include in a global model for optimum prognostic power, Figure 3 shows the results of sensitivity studies done with the WRF-CHEM model and a state-of- the –art chemistry parameterization. The sensitivity studies suggest that the process that is most important is the multi-generational chemistry of organic aerosol precursors. These findings (in greater detail) have been conveyed to NRL-Monterey (Frascati ICAP meeting, May, 2012).

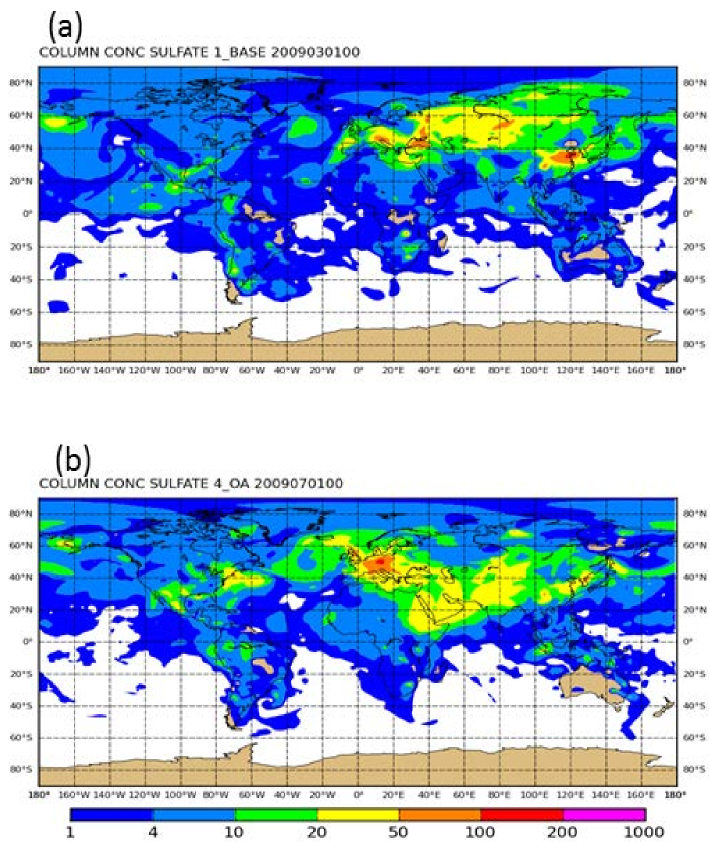

Figure 2. Sulfate column burden predicted by the NAAPS model with, (a) standard input based on widely used emissions inventories and, (b) double the standard SO_2 emission with immediate conversion to sulfate. The units are mg m^{-2}.

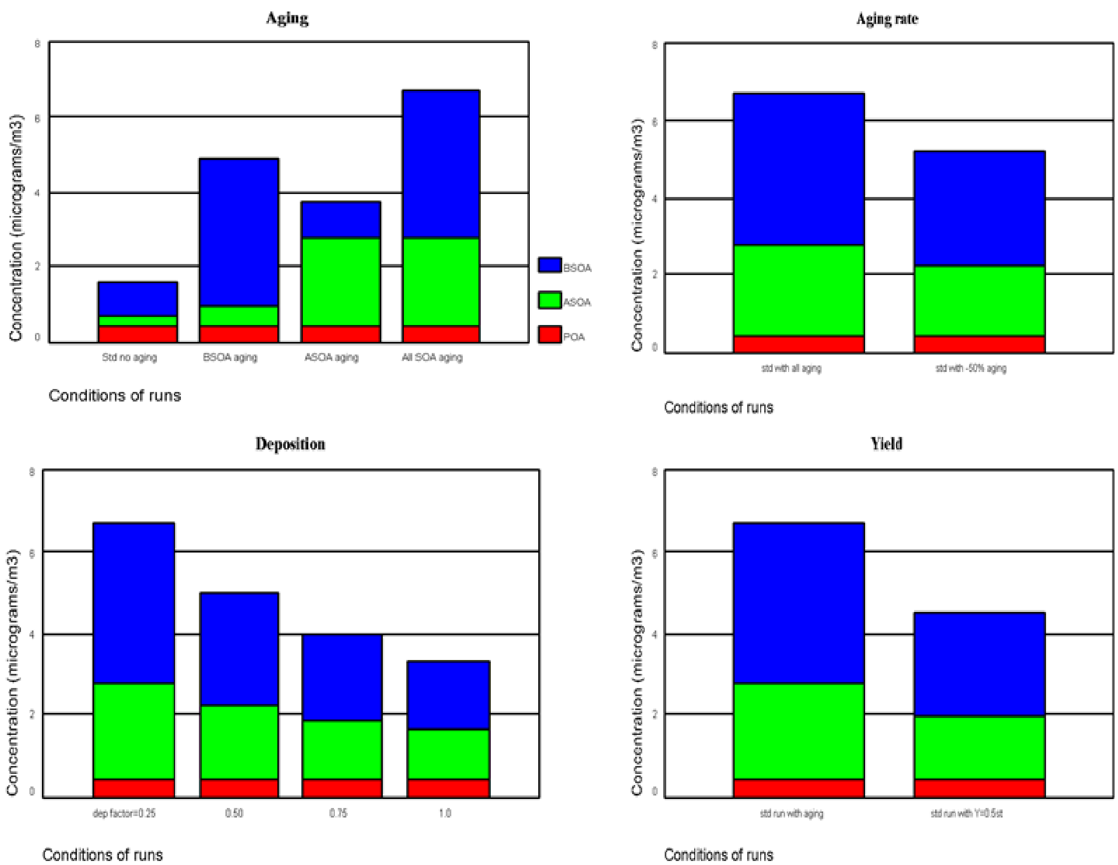

Figure 3. Results of sensitivity studies done with the WRF-CHEM model to test the relative importance of precursor aging, aging rate, reaction yield and deposition on the concentration of secondary organic aerosol. Concentrations are averages over the model domain (based on Ahmadov et al, 2012).

The last objective of this study, the aerosol optical climatology, is now well along, roughly 85% completed. We should finish it in November of this year. The data have been used in several different preliminary analyses to demonstrate their prospective value. One example is shown in Figure 4 for the CARMA II data alone. The aerosol single scattering albedo (SSA) a wavelength of 550 nm has been plotted against altitude. Given that the mean inversion height for the CARMA II study was ~ 600 m, it is quite clear that significantly more absorbent aerosol is commonly present above the MBL than in it, likely due offshore transport of biomass burning aerosol and/or pollution. Similarly, the aerosol hygroscopicity (gamma) also shows interesting vertical structure. A plot of the hygroscopic growth parameter for aerosol light scattering against altitude, this time for CARMA IV, is shown in Figure 5. The mean height of the MBL inversion for this study was ~ 450 m. Hence, as with SSA, the super inversion aerosol hygroscopicity was far more variable than that in the MBL. This can be seen quantitatively in Figure 6, in which the distribution of aerosol hygroscopicity for both the MBL and the free troposphere is show for CARMA IV. The normalized standard deviation of the mean hygroscopicity for the free troposphere is almost 6 times that for the MBL. It is also worth noting that the central value for the MBL, 0.5, suggests that ~ 50% of the aerosol mass is organic (e.g., Hegg et al, 2002). This is consistent with the source attribution for this period, based on PMF modeling (Hegg et al, 2010).

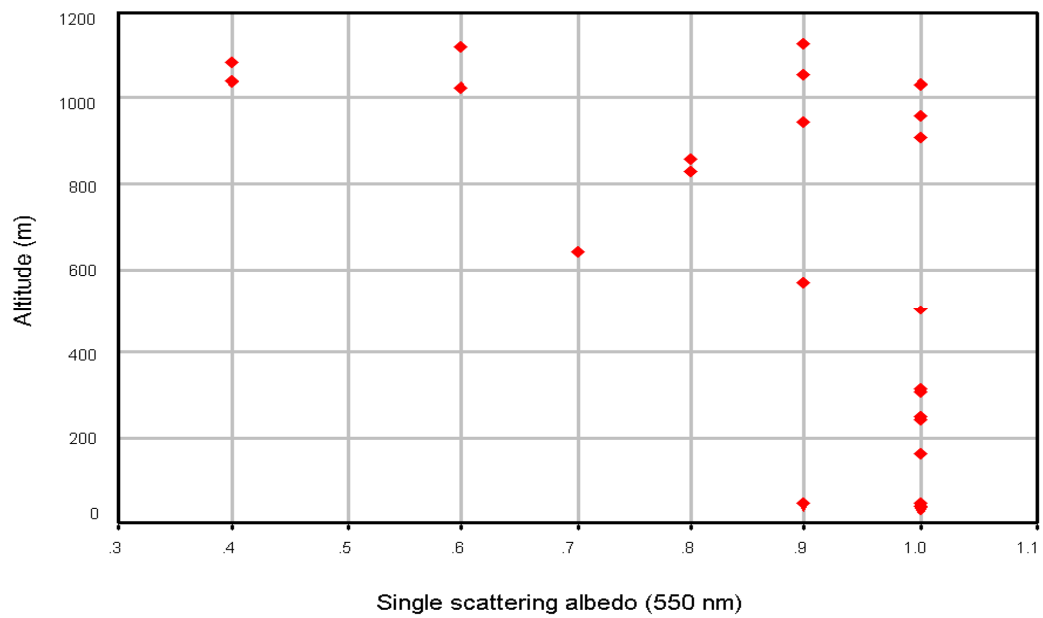

Figure 4 . The aerosol single scattering albedo (SSA) at 550 nm plotted against altitude for transects from the CARMA II study. For reference, the mean inversion height defining the top of the MBL was ~ 600 m during CARMA II.

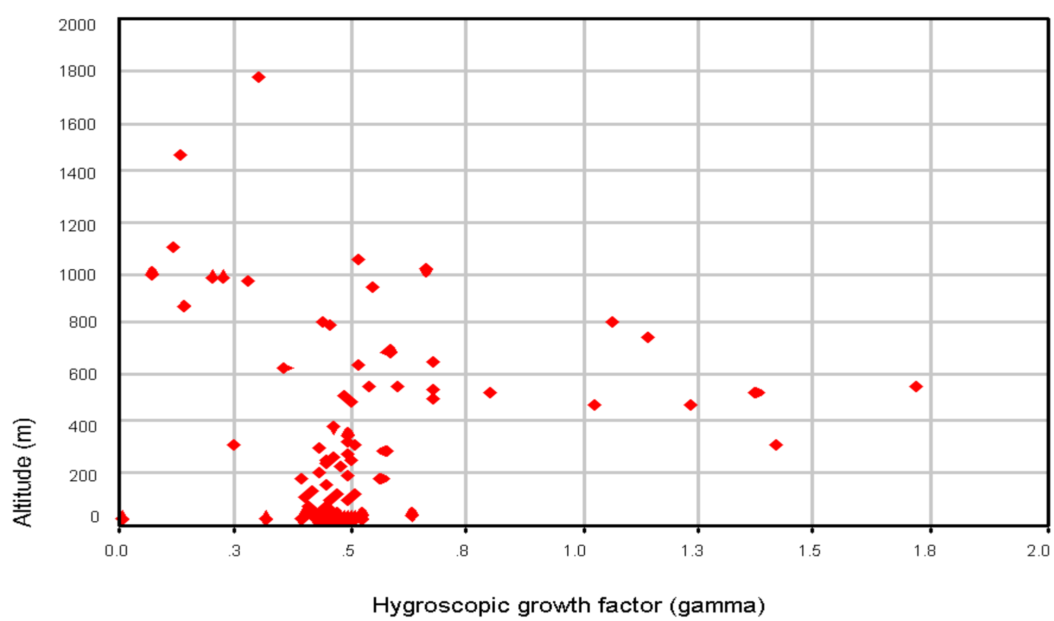

Figure 5 . The aerosol hygroscopicity plotted against altitude for transects from the CARMA IV study. The mean inversion height defining the top of the MBL was ~ 480 m during CARMA IV.

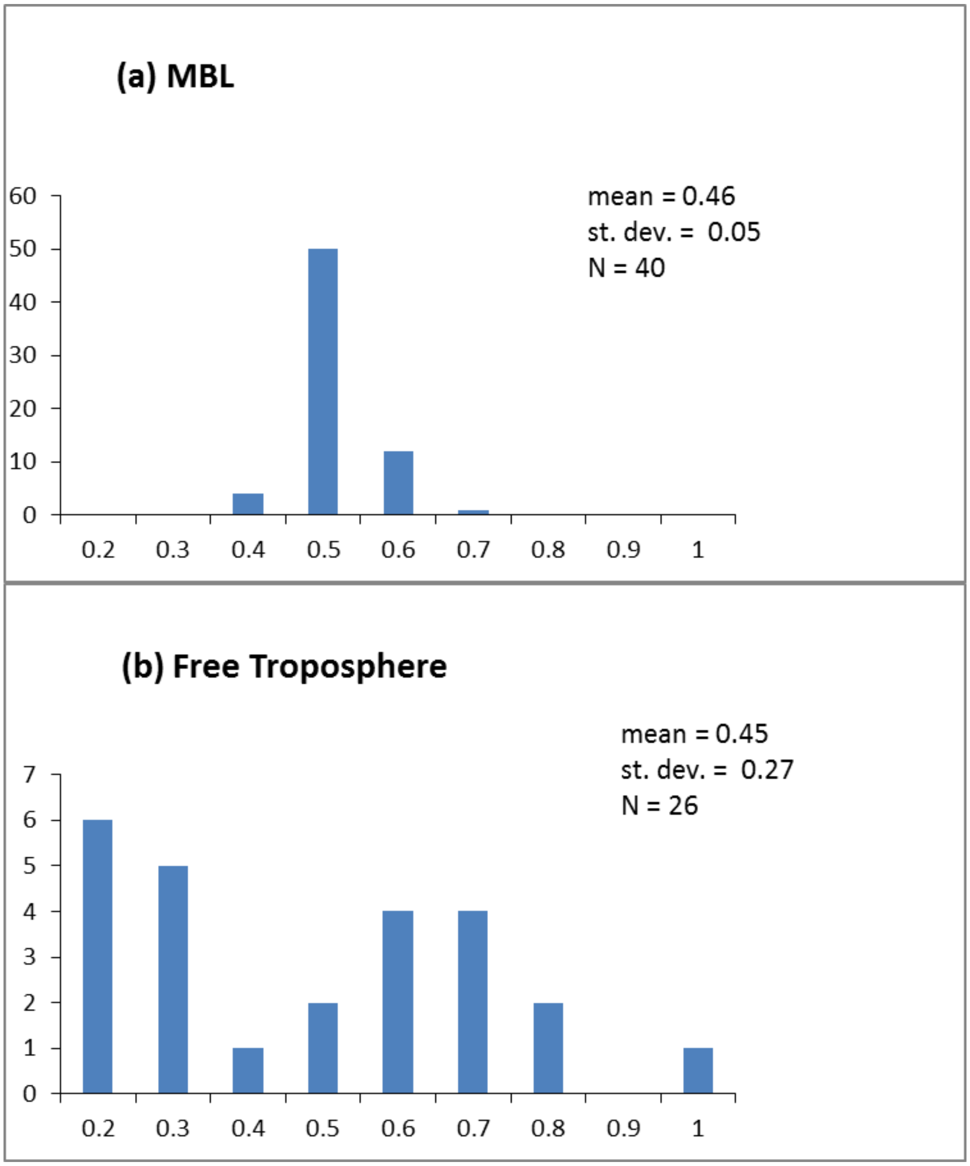

Figure 6 . Histograms of aerosol hygroscopicity for transects from the CARMA IV study. (a) in the MBL and (b) in the free troposphere.

IMPACT/APPLICATIONS

The CDNC-AMNC relationship shown in Figure 1 should have a substantial impact on the ability of remote sensing to characterize effective CCN concentrations for the important stratocumulus deck regions of the atmosphere. It should also be valuable for use in large-scale models that deal with indirect aerosol forcing. The assessment of the feasibility and methodology to employ in incorporation of SOA into the NAAPS model has the potential to significantly enhance the prognostic power of the model for AOD. The aerosol optical climatology will be a valuable resource for anyone addressing issues involving radiative transfer in and just above the MBL. It will also be a good source of input data for large scale models which encompass the marine atmosphere.

TRANSITIONS

None.

RELATED PROJECTS

These measurements are highly relevant to determination of aerosol light scattering in the MBL (and thus radiative transfer in the MBL), and CCN activity (and thus of the microphysics of MBL clouds). Furthermore, numerical transport models could now incorporate a simple relationship such as that shown in Figure 1, even for a limited domain, and thus improve their prognostic power for cloud optical properties.

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